

Heat Capacity and Thermodynamic Properties of Beryllium 1:3-Aluminate, $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$, from 15 to 390 °K

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The heat capacity of beryllium 1:3-aluminate, $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$, was determined from 15 to 380 °K and the thermodynamic properties calculated from 0 to 390 °K. The entropy at 298.15 °K was found to be 175.55 J deg⁻¹ mol⁻¹ (41.96 cal deg⁻¹ mol⁻¹).

Key Words: Beryllium 1:3-aluminate, enthalpy, entropy, Gibbs energy, heat capacity, low temperature calorimetry, thermodynamic properties.

1. Introduction

The results of the heat-capacity measurements on beryllium 1:3-aluminate,² $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$, from 15 to 380 °K presented herein have been obtained as a part of the program at the National Bureau of Standards to provide accurate thermodynamic data on "light-element" compounds. Earlier measurements on the $\text{BeO}-\text{Al}_2\text{O}_3$ system that have been reported from this laboratory are aluminum oxide (corundum), Al_2O_3 [3],³ and beryllium 1:1-aluminate (chrysoberyl), $\text{BeO} \cdot \text{Al}_2\text{O}_3$ [6]. Measurements on beryllium oxide, BeO , of relatively small crystal size have recently been completed [4] and will be reported in a future paper.

2. Sample

Lang et al. [9], reported "practically all $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$ " and "all $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$ " for the results of petrographic and x-ray examinations, respectively, on a sample prepared by fusing a mixture of 1 to 3 molar ratio of BeO to Al_2O_3 . Semi-Elements, Inc. prepared, on request, the sample accordingly for the heat-capacity measurements presented. The preparation procedure was as follows: stoichiometric amounts of dried BeO and Al_2O_3 powder were thoroughly mixed and fused in an arc furnace. To avoid contamination a fused mass was formed within a relatively large charge of the mixture so that the unfused mixture

would serve as the container for the fused portion of the material. The fused material was cooled to room temperature in about three hours, freed of any unfused material, and crushed and sieved to collect particle sizes between 0.3 and 2 mm (10 and 50 mesh) on the edge.

Some of the particles in the sample that was received appeared gray, presumably from occlusion of traces of carbon from the graphite electrodes used in the arc fusion preparation. Qualitative spectrochemical analysis of the material by the Spectrochemical Analysis Section of the National Bureau of Standards showed the limits of impurities given in table 1.

TABLE 1. Spectrochemical analysis of beryllium 1:3-aluminate ($\text{BeO} \cdot 3\text{Al}_2\text{O}_3$)^a

Element	Percentage limit	Element	Percentage limit
Ag	< 0.0001	Mg	0.001–0.01
B	0.001–0.01	Mn	0.0001–0.001
Ca	0.001–0.01	Na	0.01–0.1
Cu	< 0.0001	Si	0.01–0.1
Fe	0.01–0.1	Zr	0.001–0.01

^a Analyzed by Elizabeth K. Hubbard, Spectrochemical Analysis Section.

A petrographic (microscopic) examination of the $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$ sample was made in the Crystallography Section of the National Bureau of Standards. The crystal size was found to be on the average about 300 μ .

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²The numbers indicate base to acid anhydride ratio.

³Figures in brackets indicate references listed at the end of this paper.

Since the sample was ground for the microscopic examination, the crystal size of the original 10 to 50 mesh polycrystalline material on which the heat-capacity measurements were made is considered to be larger than 300 μ . The crystals were clear with some voids and growth defects resulting probably from too rapid cooling. There was some evidence of another phase being present, possibly up to about 5 percent. No BeO phase was detected.

X-ray diffraction examination of the BeO \cdot 3Al₂O₃ sample was also made in the Crystallography Section of the NBS. The pattern obtained is in agreement with published values for BeO \cdot 3Al₂O₃ [1, 2, 7] with some possible impurity lines close to BeO \cdot Al₂O₃ and α -Al₂O₃ but without presence of all strong lines of these substances. A few unidentified lines were also found. The patterns corresponding to graphite, BeO, and 3BeO \cdot Al₂O₃ were absent.

Optical properties of BeO \cdot Al₂O₃ and BeO \cdot 3Al₂O₃ have been found to be very similar [2, 7, 9, 10]. It seems, therefore, that the impurity phase observed petrographically in the sample investigated is probably not BeO \cdot Al₂O₃. If the undetectable BeO \cdot Al₂O₃ is present, then the excess Al₂O₃ should be observed as α -Al₂O₃ or there is an unidentified compound of BeO and Al₂O₃ of ratio greater than 1 to 3. The x-ray pattern obtained does not definitively indicate the presence of α -Al₂O₃.

Distinctive x-ray diffraction patterns have been reported for BeO \cdot Al₂O₃ and BeO \cdot 3Al₂O₃ [1, 2, 7]. Lang et al. [9], observed an unidentified phase of high refractive index in mixtures that have been heated to melting temperatures, having compositions between BeO \cdot Al₂O₃ and Al₂O₃. These investigators also found another unidentified phase in x-ray diffraction patterns of fused mixtures of composition between BeO \cdot Al₂O₃ and BeO. The major *d*-spacings observed for the latter impurity phase cannot be identified with the *d* values reported for the more recently found 3BeO \cdot Al₂O₃ phase [7]. Lang et al. [9], could not determine the presence of both of the unidentified phases in the same specimen or any similarity, if any, in the phases. They were not able to identify either phase in the same specimen by both petrographic and x-ray methods. These results seem to indicate that yet unidentified phases exist in the BeO—Al₂O₃ system or additional careful study of the known phases is needed.

Young [13] observed from x-ray measurements some disproportionation of BeO \cdot 3Al₂O₃ to BeO \cdot Al₂O₃ and Al₂O₃ after heating for two weeks at 1150 °C. This suggests that BeO \cdot 3Al₂O₃ is metastable in the range of the heat-capacity measurements obtained. Earlier, Foster and Royal [2] found BeO \cdot 3Al₂O₃ to have a stronger tendency toward crystal growth than BeO \cdot Al₂O₃ and found no evidence for BeO \cdot 3Al₂O₃ to disproportionate. The relatively rapid cooling during the preparation of the sample used in the measurements presented here has apparently preserved the material mostly, if not all, in the BeO \cdot 3Al₂O₃ phase.

Chemical analysis of the BeO \cdot 3Al₂O₃ sample was performed in the Analysis and Purification Section of the NBS. Two 0.5 g samples were dissolved in hydro-

chloric acid by heating the mixture sealed in tubes at 250 °C for 16 hr. The treatment of the resulting solution and the analysis for Be and Al were essentially the same as those described for BeO \cdot Al₂O₃ [6]. The results of the chemical analysis, summarized in table 2, show the beryllium and aluminum composition to have the theoretical stoichiometric ratio within the precision of the analytical method. The amount of graphite impurity was considered insignificant, and the sample was taken to be 100 percent BeO \cdot 3Al₂O₃ in the processing of the heat-capacity data obtained.

TABLE 2. Chemical analysis of beryllium 1:3-aluminate (BeO \cdot 3Al₂O₃)

Sample	Percentage by weight ^a		Mol ratio
	Al	Be	Al ₂ O ₃ /BeO
1	48.96	^b 2.71	3.017
2	48.93	^b 2.74	2.984
Theoretical	48.92	2.72	3.004

^a Analysis by E. J. Maienthal, Analysis and Purification Section.

^b Duplicate aliquots.

Although BeO \cdot 3Al₂O₃ is considered to be inert, the sample was handled and poured into the calorimeter vessel in a controlled-atmosphere box containing dry argon gas (dew-point of about -80 °C). The calorimeter vessel was provided with a screw-cap and gasket closure and a "pump out" tube with a valve previously described [5]. The vessel containing the sample was pumped to a high vacuum and purged with dry helium gas several times and finally sealed with 60.5 torr pressure of helium gas. The mass of sample investigated was 252.1429 g.

3. Apparatus and Method

The heat-capacity measurements on BeO \cdot 3Al₂O₃ were made in an adiabatic calorimeter similar in principle and design to that described previously [11]. The calorimeter, sample container, and the adjuvant instruments were the same, except for the bridge, as those used for the recently published measurements on BeO \cdot Al₂O₃ [6]. Description of methods and procedures used for the measurements of temperature, power, and time interval of heating, and the calibration information on the temperature scale and electrical instruments are given in the reference.

An automatic Mueller bridge, capable of resistance measurements up to almost 500 Ω to the nearest 0.00001 Ω , was used with the platinum-resistance thermometer for most of the temperature measurements. The performance of this automatic bridge was found comparable to the high-precision manually

operated Mueller bridge used exclusively in the measurements with $\text{BeO} \cdot \text{Al}_2\text{O}_3$ [6]. The bridge readings were automatically recorded on punch cards and subsequently processed on the high-speed digital computer to obtain the temperatures associated with each heating interval.

The 1961 atomic weights based on carbon 12 were used for converting the mass of sample investigated to gram formula weight basis [8]. The energy measurements were made in terms of the MKSA unit of energy, the joule. Wherever conversion to calorie was made, the following relation was used:

$$1 \text{ defined calorie} = 4.1840 \text{ J.}$$

4. Heat-Capacity Measurements and Results

Measurements totaling 147 heat-capacity "points" were obtained on the calorimeter vessel plus sample from about 15 to 390 °K and 87 points were obtained subsequently on the empty vessel in the same temperature range. Smoothed values of heat capacity at closely and regularly spaced integral temperatures were calculated for the two series of measurements using high-speed digital computer methods. The heat capacity of the sample was then obtained by differencing the smoothed values from the two series of measurements at the corresponding regular temperatures. The values of heat capacity for the $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$ below 15 °K were obtained by extrapolating the smoothed experimental values at the lower temperatures in accordance with a T^3 relation.

Briefly, the procedures used in analyzing the experimental observations were as follows. Since the data could not be adequately represented over the complete temperature range in terms of a single heat-capacity equation, a number of polynomials of varying degrees of complexity were fitted by the method of least squares to the data over different temperature ranges. To avoid curvature corrections the analysis was formulated according to the relation:

$$Q_{T_2, T_1} = H(T_2) - H(T_1) = \int_{T_1}^{T_2} C(T) dT,$$

where Q_{T_2, T_1} is the input energy, T_1 and T_2 the corresponding initial and final temperatures, respectively, for each of the points, and $C(T)$ is the polynomial for which the coefficients were obtained by the least-squares method. Three or four polynomials were selected from the group on the basis of their fit over the complete temperature range of observations and for their agreement where the temperatures overlap. The polynomials were evaluated at closely spaced regular temperatures and joined at temperatures where

the values of heat capacity and their first and second derivatives showed the best agreement. A numerical smoothing analysis was performed on these tabular values using the computer to test the smoothness of the joining process. Since the values were adequately smooth, they were changed insignificantly by the computer smoothing program.

The deviations of the observed values of heat capacity for the calorimeter-vessel-plus-sample and for the empty-vessel measurements from the final smoothed tabular values for the two series of measurements are plotted in figures 1 and 2, respectively. Except below about 50 °K, all of the observations are well within the ± 0.1 percent limit of the net heat capacity of the $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$ sample.

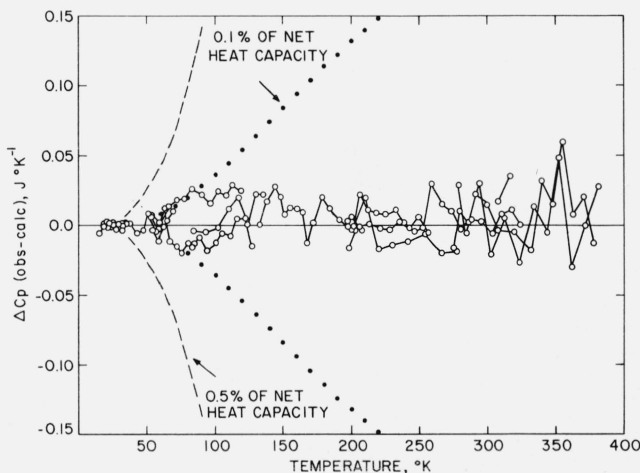


FIGURE 1. Deviations of the heat-capacity measurements on calorimeter vessel plus beryllium 1:3-aluminate, $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$.

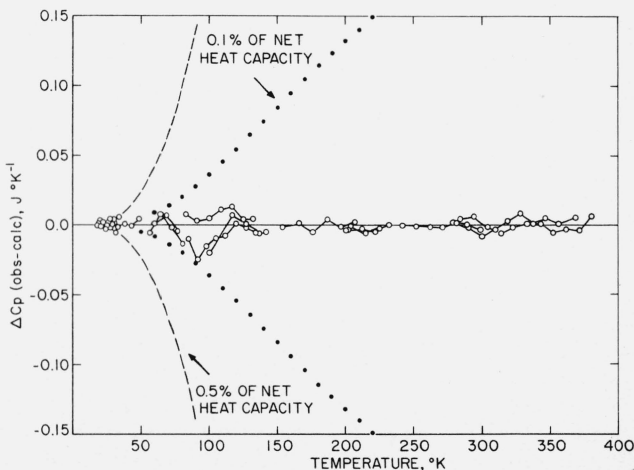


FIGURE 2. Deviations of the heat-capacity measurements on the empty calorimeter vessel used with beryllium 1:3-aluminate, $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$.

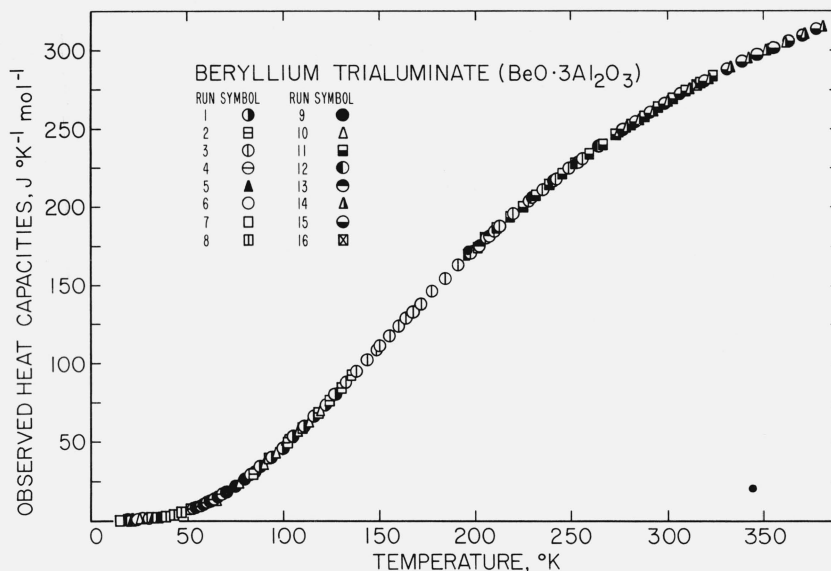


FIGURE 3. Observed values of the molal heat capacity of beryllium 1:3-aluminate, $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$, as a function of the temperature.
(Numerical values are given in table 3.)

The "observed molal heat capacity" is shown in figure 3 and listed in table 3 in the chronological order of the measurements made. These values were obtained by subtracting the smoothed heat capacity of the empty vessel from that of the calorimeter vessel plus sample at the observed temperatures (mid-temperatures of the heating intervals). The heat-capacity values of the empty vessel at the corresponding observed temperatures were obtained by interpolation in the table of smoothed values described earlier. Corrections were made for the heat capacity of helium gas and for curvature wherever significant and the net heat capacity converted to the molal observed values.

The final smoothed values of heat capacity of $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$ given in table 4 were obtained, however, by differencing at corresponding temperatures the smoothed values described earlier for the two series of measurements and correcting for the contribution of the helium exchange gas wherever significant. The smoothness of the resulting tabular values was checked on the computer and converted to molal values.

The smoothed values at the lower temperatures were plotted as C/T versus T^2 and extrapolated linearly to 0 °K to obtain values below 15 °K, the lower temperature limit of the experimental measurements. The slope of the line used was $4.025 \times 10^{-5} \text{ J deg}^{-4} \text{ mol}^{-1}$, which corresponds to an effective Debye characteristic temperature of 936 °K (assuming $C_V = C_P$).

5. Reliability of the Results

In addition to the imprecision of the measurements shown in figures 1 and 2, the purity of the sample, the accuracy of calibration of the instruments used, and the possible sources of systematic errors were examined to estimate the uncertainties in the final values of heat capacity of $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$. The estimated uncertainty in the values given in table 4 between 80 and 390 °K is ± 0.1 percent. Below about 80 °K, the uncertainty increased, because of the decrease in the contribution of the sample to the gross heat capacity. At 300 °K the heat capacity of the sample was 81 percent of the gross; at 80 °K, 46 percent; at 50 °K, 29 percent; 20 °K, 14 percent; and at 13 °K, only 10 percent. In addition, the sensitivity dR/dT of the platinum resistance thermometer began to decrease significantly below about 40 °K. Considering these factors, the estimated uncertainty is ± 0.2 percent at 50 °K, ± 1 percent at 20 °K, and ± 3 percent at 13 °K.

6. Thermodynamic Functions and Discussion of the Results

The thermodynamic functions for $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$ were derived, using their usual relations with respect to heat capacity, from the smoothed tabular values. The thermodynamic relations were evaluated on the com-

TABLE 3. Observed heat capacity of beryllium 1:3-aluminate ($\text{BeO} \cdot 3\text{Al}_2\text{O}_3$)

Gram molecular wt = 330.8952 g		1 cal = 4.1840 J		$T^\circ\text{K} = 273.15 + t^\circ\text{C}$	
T	C_p	T	C_p	T	C_p
RUN 1		RUN 6—Con.		RUN 11—Con.	
$^\circ\text{K}$	$J/\text{deg-mol}$	$^\circ\text{K}$	$J/\text{deg-mol}$	$^\circ\text{K}$	$J/\text{deg-mol}$
82.8203	28.964	23.3504	0.532	295.3757	263.122
88.0822	34.227	26.8876	0.827	302.7188	268.501
93.6560	40.027	30.7964	1.270	309.9404	273.633
99.4640	46.435			317.0493	278.493
105.1819	53.076			324.0556	283.101
110.8937	59.946	RUN 7		RUN 12	
116.2799	66.639				
121.5671	73.341	14.7135	0.107	199.6885	172.558
126.9319	80.234	19.0305	0.279	209.5623	183.776
RUN 2		21.4966	0.414	220.4510	195.579
		24.9047	0.654	230.4099	205.954
		28.9755	1.047	241.5414	217.011
83.4430	29.619	RUN 8		253.8218	228.599
93.1378	39.520			265.6168	239.092
102.3438	49.768			277.0019	248.759
110.0670	58.972			RUN 13	
117.2945	67.939				
123.8566	76.279	25.6653	0.718	198.0565	170.655
129.9306	84.191	29.7872	1.147	201.9686	175.189
135.6114	91.641	33.8800	1.737	207.2721	181.202
RUN 3		38.4138	2.618	RUN 14	
		42.7876	3.715		
		46.9192	5.045	275.0636	247.116
		51.3870	6.817	278.9339	250.319
		56.0607	9.017	285.8983	255.860
132.6454	87.716	RUN 9		293.6534	261.865
138.1908	95.032			301.7499	267.778
143.4155	101.938			311.7800	274.892
148.3858	108.477			322.8485	282.271
150.3864	111.090	53.0258	7.546	333.3522	288.997
155.0609	117.220	57.5249	9.794	343.2091	294.909
159.5568	123.071	61.8323	12.339	352.1076	300.070
163.8977	128.677	66.0923	15.097	361.4226	305.043
167.4038	133.143	70.4018	18.206	371.2744	310.239
171.4439	138.297	74.9699	21.849	380.9849	315.212
177.6536	146.111	79.9109	26.210	RUN 15	
184.5438	154.587	85.1544	31.264		
191.1511	162.544	RUN 10		277.8938	249.497
197.5135	170.039			284.1183	254.451
199.9364	172.860			290.9840	259.825
206.0172	179.794			298.9705	265.799
212.9353	187.490	53.4833	7.764	307.2420	271.725
220.6606	195.831	58.9604	10.617	319.3254	279.963
228.1316	203.644	64.5559	14.102	331.2745	287.670
235.3762	210.991	70.6195	18.435	339.1056	292.530
242.4181	217.892	77.0761	23.717	346.9111	297.086
249.2865	224.433	83.3997	29.590	354.6113	301.477
255.9950	230.597	89.8264	36.063	362.3108	305.570
RUN 4		96.1066	42.727	369.8374	309.529
		102.1024	49.512	377.2870	313.293
		107.8188	56.257	RUN 16	
		113.3406	63.011		
		119.0752	70.194	307.6850	272.065
54.4277	8.188	RUN 11		315.9765	277.803
57.6286	9.833				
60.6838	11.611				
64.0585	13.748	196.3894	168.737		
68.1789	16.598	200.5662	173.578		
RUN 5		206.1477	179.973		
		212.0283	186.512		
21.9906	0.446	218.3673	193.399		
25.3842	0.691	224.7804	200.188		
28.5606	1.002	231.5555	207.161		
32.1099	1.452	238.6756	214.253		
35.5483	2.025	245.6172	220.957		
RUN 6		252.3915	227.307		
		259.3612	233.673		
		266.5257	239.939		
		273.6103	246.048		
18.4505	0.250	280.6552	251.690		
21.0068	0.381	287.9167	257.439		

TABLE 4. Thermodynamic functions for beryllium 1:3-aluminate ($\text{BeO} \cdot 3\text{Al}_2\text{O}_3$) solid phase.

Gram molecular wt = 330.8952g						
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TABLE 4. Thermodynamic functions for beryllium 1:3-aluminate ($\text{BeO} \cdot 3\text{Al}_2\text{O}_3$) solid phase.

Gram Molecular wt = 330.8952 g 1 cal = 4.1840 J
 $T^\circ\text{K} = 273.15 + t^\circ\text{C}$

T	C_p°	$(H_T^\circ - H_0^\circ)$	$(H_T^\circ - H_0^\circ)/T$	$(S_T - S_0^\circ)$	$-(G_T^\circ - H_0^\circ)$	$-(G_T^\circ - H_0^\circ)/T$
$^\circ\text{K}$	J/deg-mol	J/mol	J/deg-mol	J/deg-mol	J/mol	J/deg-mol
320.00	280.46	40077.	125.24	194.85	22275.	69.609
325.00	283.72	41488.	127.65	199.22	23260.	71.569
330.00	286.90	42914.	130.04	203.58	24267.	73.537
335.00	290.00	44356.	132.41	207.92	25296.	75.510
340.00	293.02	45814.	134.75	212.24	26346.	77.489
345.00	295.96	47287.	137.06	216.54	27418.	79.473
350.00	298.82	48774.	139.35	220.81	28512.	81.461
355.00	301.60	50275.	141.62	225.07	29626.	83.454
360.00	304.32	51789.	143.86	229.31	30762.	85.451
365.00	306.98	53318.	146.08	233.53	31919.	87.450
370.00	309.59	54859.	148.27	237.72	33097.	89.452
373.15	311.21	55837.	149.64	240.35	33850.	90.715
375.00	312.15	56413.	150.44	241.89	34296.	91.457
380.00	314.68	57981.	152.58	246.04	35516.	93.464
385.00	317.19	59560.	154.70	250.17	36757.	95.472
390.00	319.69	61152.	156.80	254.28	38018.	97.482

H_0° and S_0° apply to the reference state of the solid at zero $^\circ\text{K}$.

puter by numerical integration procedures with four-point Lagrangian integration coefficients [12]. The thermodynamic functions are given in table 4 from 0 to 390 $^\circ\text{K}$.

Although x-ray diffraction measurements on $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$ have been reported [1, 2, 7], no information on the crystal structure of the material has been found. The room temperature density (3.73 g cm^{-3} [2]) of $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$ indicates that the crystal is more loosely bound on the average than the parent oxides, BeO and Al_2O_3 . The molal volume, $V(\text{BeO} \cdot 3\text{Al}_2\text{O}_3)$, of $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$ is larger (4.03%) than the sum of the molal volumes, $V(\text{BeO}) + 3V(\text{Al}_2\text{O}_3)$, of BeO and Al_2O_3 , respectively. The molal volume of $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$ is also larger (3.56%) than the sum of the molal volumes, $V(\text{BeO} \cdot \text{Al}_2\text{O}_3) + 2V(\text{Al}_2\text{O}_3)$, of $\text{BeO} \cdot \text{Al}_2\text{O}_3$ and Al_2O_3 , respectively. The molal volume of $\text{BeO} \cdot \text{Al}_2\text{O}_3$ is likewise larger (1.21%) than the volume of equimolal mixture of BeO and Al_2O_3 .

A comparison (fig. 4) of the heat capacity of $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$ with those of the corresponding molar mixtures of BeO and Al_2O_3 and of $\text{BeO} \cdot \text{Al}_2\text{O}_3$ and Al_2O_3 shows a positive deviation throughout the temperature range (15 to 390 $^\circ\text{K}$), with the deviations increasing with decrease in temperature. A similar positive deviation from "additivity" of the heat capacity was found with $\text{BeO} \cdot \text{Al}_2\text{O}_3$ [6] (fig. 4) and with many mixed metal oxides reported in the literature. Also, with the exception of a few, where the density data may possibly be in error, a positive deviation from the additivity of the molal volumes was found at room temperature for the same mixed metal oxides. For substances at temperatures below the classical heat capacity limit, this correspondence is reasonable since a more open structure yields a "softer" force constant and hence, a lower Debye characteristic temperature. Further comparison of heat capacity and volume over a wide range of temperature is in progress, but unfortunately

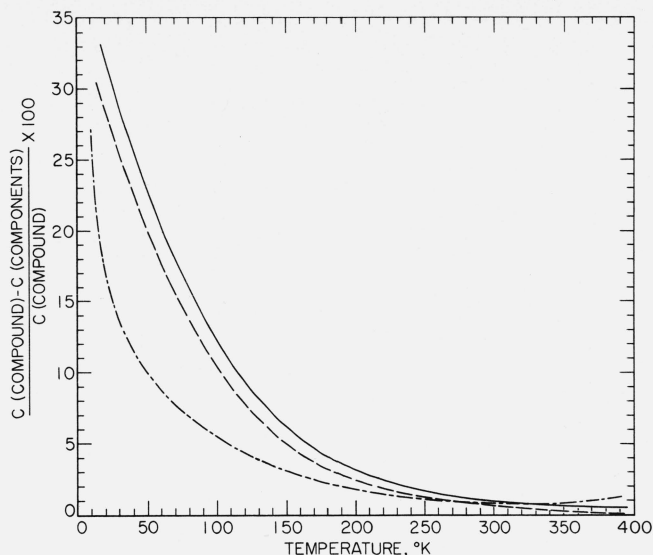


FIGURE 4. Comparison of the additivity of heat capacities:

— $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$ versus $\text{BeO} + 3\text{Al}_2\text{O}_3$
 - - $\text{BeO} \cdot 3\text{Al}_2\text{O}_3$ versus $\text{BeO} \cdot \text{Al}_2\text{O}_3 + 2\text{Al}_2\text{O}_3$
 - · - $\text{BeO} \cdot \text{Al}_2\text{O}_3$ versus $\text{BeO} + \text{Al}_2\text{O}_3$

relatively few thermal expansion data seem to be available for the range below room temperature.

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